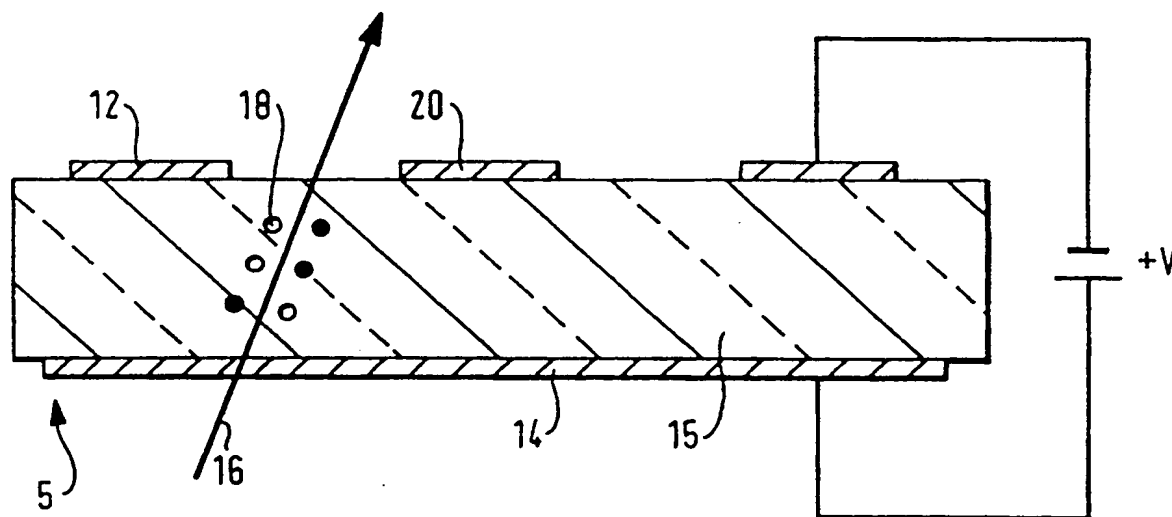




## INTERNATIONAL APPLICATION PUBLISHED UNDER THE PATENT COOPERATION TREATY (PCT)

<b>(51) International Patent Classification <sup>6</sup>:</b> <b>G01T 1/26, 3/08</b>	<b>A1</b>	<b>(11) International Publication Number:</b> <b>WO 97/00456</b> <b>(43) International Publication Date:</b> 3 January 1997 (03.01.97)
<b>(21) International Application Number:</b> PCT/GB96/01357 <b>(22) International Filing Date:</b> 6 June 1996 (06.06.96) <b>(30) Priority Data:</b> 9512057.2 14 June 1995 (14.06.95) GB <b>(71) Applicant (for all designated States except US):</b> IMPERIAL COLLEGE OF SCIENCE, TECHNOLOGY & MEDICINE [GB/GB]; Exhibition Road, London SW7 2AZ (GB). <b>(72) Inventors; and</b> <b>(75) Inventors/Applicants (for US only):</b> HASSARD, John, Francis [GB/GB]; Selkirk Hall, Princes Gardens, London SW7 1LU (GB). GODDARD, Antony, John, Hudson [GB/GB]; 20 Maze Road, Kew, Surrey TW9 3DE (GB). <b>(74) Agents:</b> MAGGS, Michael, Norman et al.; Kilburn & Strode, 30 John Street, London WC1N 2DD (GB).		<b>(81) Designated States:</b> AL, AM, AT, AU, AZ, BB, BG, BR, BY, CA, CH, CN, CZ, DE, DK, EE, ES, FI, GB, GE, HU, IL, IS, JP, KE, KG, KP, KR, KZ, LK, LR, LS, LT, LU, LV, MD, MG, MK, MN, MW, MX, NO, NZ, PL, PT, RO, RU, SD, SE, SG, SI, SK, TJ, TM, TR, TT, UA, UG, US, UZ, VN, ARIPO patent (KE, LS, MW, SD, SZ, UG), Eurasian patent (AM, AZ, BY, KG, KZ, MD, RU, TJ, TM), European patent (AT, BE, CH, DE, DK, ES, FI, FR, GB, GR, IE, IT, LU, MC, NL, PT, SE), OAPI patent (BF, BJ, CF, CG, CI, CM, GA, GN, ML, MR, NE, SN, TD, TG).  <b>Published</b> <i>With international search report.</i>

(54) Title: NEUTRON DETECTOR



## (57) Abstract

A neutron detector (5, 10) comprises a diamond detector element (15, 40) doped with boron. Boron-doped diamond substantially improves the rate of neutron detection due to the large amount of pair production, and is extremely mechanically and thermally robust. In one embodiment (15) of the invention, the detector is planar. A second embodiment (10) uses a series of ridges (40) and improves the response rate still further; the incident neutron energy, position and time of incidence upon the detector is also enhanced in comparison with prior art detectors. The detector finds particular application in the field of slow (thermal) neutron detection, but is nonetheless useful in fast neutron spectroscopy.

**FOR THE PURPOSES OF INFORMATION ONLY**

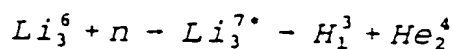
Codes used to identify States party to the PCT on the front pages of pamphlets publishing international applications under the PCT.

AM	Armenia	GB	United Kingdom	MW	Malawi
AT	Austria	GE	Georgia	MX	Mexico
AU	Australia	GN	Guinea	NE	Niger
BB	Barbados	GR	Greece	NL	Netherlands
BE	Belgium	HU	Hungary	NO	Norway
BF	Burkina Faso	IE	Ireland	NZ	New Zealand
BG	Bulgaria	IT	Italy	PL	Poland
BJ	Benin	JP	Japan	PT	Portugal
BR	Brazil	KE	Kenya	RO	Romania
BY	Belarus	KG	Kyrgyzstan	RU	Russian Federation
CA	Canada	KP	Democratic People's Republic of Korea	SD	Sudan
CF	Central African Republic	KR	Republic of Korea	SE	Sweden
CG	Congo	KZ	Kazakhstan	SG	Singapore
CH	Switzerland	LI	Liechtenstein	SI	Slovenia
CI	Côte d'Ivoire	LK	Sri Lanka	SK	Slovakia
CM	Cameroon	LR	Liberia	SN	Senegal
CN	China	LT	Lithuania	SZ	Swaziland
CS	Czechoslovakia	LU	Luxembourg	TD	Chad
CZ	Czech Republic	LV	Latvia	TG	Togo
DE	Germany	MC	Monaco	TJ	Tajikistan
DK	Denmark	MD	Republic of Moldova	TT	Trinidad and Tobago
EE	Estonia	MG	Madagascar	UA	Ukraine
ES	Spain	ML	Mali	UG	Uganda
FI	Finland	MN	Mongolia	US	United States of America
FR	France	MR	Mauritania	UZ	Uzbekistan
GA	Gabon			VN	Viet Nam

### Neutron Detector

This invention relates to a neutron detector.

5 The problem of how to detect neutrons has concerned workers in the fields of reactor physics, health physics and academic research for some time. The neutrons, which are uncharged particles, can only be detected by their interaction with charged particles such as protons or  
10 light nuclei. One such reaction is:



The problem with using lithium to detect neutrons is that it cannot be made into a solid state detector, predominantly because of its high volatility. Other  
15 elements that have also been used to attempt to detect neutrons and use a similar mechanism to the neutron-to-triton mechanism above are He<sup>3</sup> (which must also be used in gaseous form), N<sup>14</sup>, S<sup>32</sup> and Cl<sup>35</sup>. The spatial, temporal and  
20 energy resolutions of known neutron detectors remain substantially poorer than corresponding charged particle detectors. Measuring the energy of an individual neutron is extremely difficult, and even simply detecting the presence of neutrons poses problems which known detector materials have not adequately solved. Furthermore, it is  
25 extremely difficult using known neutron detectors to measure the time of incidence of a neutron, a problem which is related to the energy measurement if one considers the time-of-flight. Further, in the non-destructive testing technique known as neutron  
30 radiography, it is currently impossible to devise a neutron detector which has both a high spatial resolution and a very fast dynamic response.

It is an object of the present invention to provide a neutron detector whose detection properties are superior to those of the prior art.

5 According to the present invention there is provided a neutron detector comprising a plurality of boron-doped diamond detector elements having generally parallel sides, the sides carrying readout electrodes.

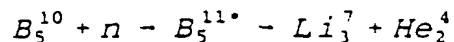
10 The combination of the shape and constitution provides a detector that is relatively cheap to manufacture, is highly sensitive, has an extremely fast response time (less than 50 picoseconds), and provides very accurate  
15 positioning information without significant cross-talk between channels. The energy and time of incidence of the neutron are also measurable with some precision. In contrast to a planar geometry, the parallel sides of the detector allow excellent containment of the products of  
20 interaction between the boron-doped diamond and the neutrons.

In addition, this topography has a directional response which is dependent upon the aspect ratio of the height of the parallel walls to the gap between them. By varying  
25 this ratio, the detector's response may be made more or less dependent on the angle of incidence of the incident neutrons.

Boron acts as a substitutional acceptor. The diamond  
30 lattice is able to accept an extremely large concentration of boron, which has a huge capture cross section for neutrons (approximately  $7.5 \times 10^{-22}$  cm<sup>2</sup>) but a relatively small scattering cross section ( $4 \times 10^{-24}$  cm<sup>2</sup>). In addition, a large amount of energy is released upon  
35 neutron capture, allowing ready detection over a wide

range of energies.

The detection occurs through the reaction:



5 and the alpha particle is readily detected by its production of electron-hole pairs which are collected by the readout electrodes. Both reaction products, however, are capable of generating an easily resolvable signal against which the presence of other radiations can be discriminated. B<sup>10</sup> occurs at the level of about 20% of naturally occurring boron, but may be extracted to produce essentially isotopically pure B<sup>10</sup>.  
10 Alternatively, doping with B<sup>11</sup> may be considered.

15 Since diamond is the subject of the boron doping, other advantages over the prior art such as excellent mechanical and thermal robustness together with radiation hardness are achieved. Boron triflouride gas filled counters will detect neutrons using a similar reaction to  
20 that above, but suffer from effects of dampness or vibration and require regular maintenance.

25 The boron concentration is preferably 10<sup>20</sup> atoms cm<sup>-3</sup> or less, for example less than 10<sup>17</sup> atoms cm<sup>-3</sup>. Diamond doped with about 10<sup>20</sup> atoms cm<sup>-3</sup> of boron is known as type IIB diamond and appears slightly blue owing to absorption in the violet and blue regions of the visible spectrum. Once the boron concentration rises substantially above 10<sup>18</sup> atoms cm<sup>-3</sup>, increased dark current leakage (noise) occurs  
30 and the diamond becomes a semiconductor. There is a trade off between increased noise in the detector at high concentrations of boron and reduced sensitivity to incident neutrons (due to reduced total integrated cross

section) at lower concentrations.

Preferably, the detector element is formed by a growth method including hot filament chemical vapour deposition (HFCVD) or microwave or radio frequency plasma growth chemical vapour deposition. The quantity of boron may be controlled to quite precise values, for example by employing diborane gas ( $B_2H_6$ ) or boric acid in acetone.

Alternatively, the boron may be ion-implanted into type IIA diamond. This is most preferably carried out using an ion concentration of  $10^{14}$  atoms  $cm^{-2}$  and ion beam energy of 40 keV.

The present invention can be put into practice in various ways which will now be described by way of example with reference to the accompanying drawings in which :-

Figure 1 shows a double logarithmic plot of capture cross-section against neutron energy for boron-doped diamond;

Figure 2 shows a sectional view of a planar neutron detector according to a first embodiment of the present invention;

Figure 3 shows a perspective view of a ridge detector for detecting neutrons according to a second embodiment of the present invention; and,

Figure 4 shows a partial cross section along the line P-P' of Figure 3.

Figure 1 indicates the manner of interaction of neutrons

with boron. When the incident neutron energy is 0.01 eV, the capture cross section is  $1.1 \times 10^{-21} \text{ cm}^2$ , at 1 eV it is  $1 \times 10^{-22} \text{ cm}^2$ , and at 1 keV it is  $4 \times 10^{-24} \text{ cm}^2$ . The total capture cross section of a neutron in boron is thus inversely proportional to the velocity of the neutron, as is indicated by the negative slope of the log-log plot of Figure 1. A further remarkable feature of boron is the lack of a resonant peak in the cross-section vs. energy plot, suggesting that the signal produced is a linear function of neutron energy.

Diamond may be doped with boron using a number of well-known methods. One technique is to grow a crystal epitaxially either in a microwave/radio frequency plasma, or use hot filament chemical vapour deposition (HFCVD). Alternatively, a pre-formed type IIA diamond may be ion-implanted with boron, using a typical dose of  $10^{14} \text{ ions cm}^{-2}$  and beam energy of around 40 keV. The activation energy of a heavily doped diamond sample ( $10^{20} \text{ atoms cm}^{-3}$ ) is approximately 2 meV. At this level of doping, the dark current noise - that is, the level of "background" current produced even when there are no incident neutrons - is dramatically increased. For this reason, there is a trade off whereby increasing the doping raises the sensitivity but increases the dark current.

In the growth methods outlined above, the quantity of boron is controlled in a straightforward manner by using gases such as diborane ( $\text{B}_2\text{H}_6$ ) or boric acid in acetone.

Figure 2 shows a planar neutron detector 5 comprising a flat sheet 15 made of boron-doped diamond. The doping may be by  $\text{B}^{10}$  or  $\text{B}^{11}$ . This sheet 15 has thin gold electrode coatings 12, 14 on its upper and lower surfaces. The upper electrode coating 12 comprises a plurality of

parallel readout strips which are aligned in a direction perpendicular to the plane of the paper in the Figure, and the lower electrode coating 14 comprises a further plurality of readout strips aligned in a direction parallel with the plane of the paper. A large potential difference  $V$  is maintained between the electrode coatings.

A neutron following a path 16 through the boron-doped diamond produces excited boron atoms which rapidly decay into stable lithium atoms and alpha particles. These in turn produce electron-hole pairs 18,20, which separate under the influence of the electric field and induce a charge on the readout strips. The energy of the neutron can be determined by the amount of charge which is collected, and its position by the intersection of the upper and lower strips receiving the largest induced charges. High precision is obtained because of the large number of electron-hole pairs produced: for example, a 1 MeV alpha particle produced during the nuclear reaction will in turn produce in excess of 100,000 electron-hole pairs.

Using boron-doped diamond confers significant advantages over prior art planar neutron detectors in terms of increased sensitivity, ability to detect larger numbers of neutrons, and improved energy and time resolution. For example, undoped diamond has a relatively poor charge collection efficiency due to limits imposed by the charge intrinsic lifetime within the diamond. Nonetheless, it is sometimes advantageous to employ less heavily boron-doped diamond in order to limit the otherwise unmanageable count rate of the detector.

In order further to improve the detector characteristics,



however, the ridge arrangement of Figures 3 and 4 may be employed. Here, the detector comprises a boron-doped diamond substrate 30 having, on one surface, a plurality of parallel etched boron-doped diamond ridges 40. On one side of each ridge there is a positive readout electrode 50, and on the other side a negative electrode 60. These are preferably conductors, but could instead be of a high-conductivity doped semiconductor material.

In use, the detector is positioned in line with a source of neutrons 70 to be detected. If it is desired to detect fast neutrons, the substrate is aligned substantially normal to the direction of the neutron beam. An individual neutron passing into one of the ridges creates lithium atoms and alpha particles which in turn produce electron-hole pairs. These rapidly migrate to the electrodes 50,60 by virtue of the potential difference which is maintained between them and which is of order  $1 \text{ V } \mu\text{m}^{-1}$  in the "C" direction. Charge is thereby induced on the electrodes, this charge being read off by readout devices (not shown) at the ends of the ridges. Once again, the large numbers of electron-hole pairs produced are advantageous, and can further be registered with excellent noise discrimination in the present embodiment.

The substrate and ridges may preferably be grown using one of the techniques outlined above. The ridges may either be grown with the substrate, or they may be etched (for example with an excimer laser). The electrodes 50,60 may be of any suitable ohmic material, such as gold, platinum, titanium, chromium and so on. Standard deposition techniques may be used to apply the metal as a thin coating to the sides of the ridges. Typically, the device may be made by etching the ridges, depositing the material, and then polishing the top surface.

It will be appreciated from Figure 4 that the sensitivity of the device shown can be increased by making the value of D (or the height of the ridges) larger. The greater the height of the ridges, the larger the amount of material which a neutron has to pass through, thereby increasing the number of interaction products within the device. The readout speed and charge collection efficiency is determined substantially by the width C of each of the ridges. Depending upon the particular application, the value of C may be as little as a few micrometers, and the value of D 100 micrometres or more, preferably in excess of 200 micrometres. Greater thicknesses provide greater efficiency as they increase the integrated boron cross-section faced by the incoming neutron. The signal-to-noise ratio is large, as there is negligible cross-talk between signals emanating from individual ridges. This is because the leakage current is low which in turn minimises shot noise. The associated read-out electronics also contribute little noise even when the signal is integrated over very short time periods (e.g. between 10 and 5000 nanoseconds). A typical substrate depth is around 100 micrometres, sufficiently thick to support the ridges and to be free-standing without requiring an additional supporting base. Ideally, the substrate and the ridges are together formed from a single wafer of material.

The directionality of the response of the ridge-shaped detector may be tailored to suit the application for which the detector is to be used. This is because the sharpness of response as a function of angle depends upon both the aspect ratio  $B/C$  in Figure 4, together with the "coverage" defined as the ratio  $C/(C+A)$ .

The impedance of the readout devices (not shown) at the

end of the ridges is preferably matched with the impedance of the electrodes 50,60, thereby increasing readout speed and reducing signal losses.

5 In order further to improve the neutron detecting capability of the detector, the spaces between the ridges may be filled with a plastics material, or other absorber.

10 In a further embodiment (not shown) a further parallel set of ridges, orthogonal to the first set, is provided on the lower surface of the substrate 30.

The ridge shaped neutron detector described above can  
15 provide extremely rapid charge readout, probably within 35 ps and certainly within 50 ps. These readout speeds cannot currently be achieved for any single pulse detector of comparable sensitivity and positional accuracy. In addition, the positional resolution is  
20 better than 20  $\mu\text{m}$  (and probably better than 10  $\mu\text{m}$ ); resolution is determined by the size of the ridge top C in Figure 3.

The signal detected by the electrodes may be read out by  
25 any conventional readout electronics. In one arrangement, the pulses of electric charge deposited by the nuclear process involved may be detected as a current (in dosimetry applications, for example). Alternatively, some applications may require the  
30 detection of single neutrons (for example in radiological applications), and this may be achieved by means of suitable electronics running in charge mode. It is a particular novel feature of the detector described above that it may either be operated as a dosimeter, or as a  
35 single neutron detector, according to application.

A 1 cm<sup>3</sup> array of diamond doped with 10<sup>20</sup> atoms cm<sup>-3</sup> of boron will have a total capture cross section of 7.5x10<sup>-22</sup> cm<sup>2</sup>. When a flux of thermal (slow) neutrons having an energy of 25 meV is incident upon this, calculations suggest that at least 7.5% of the incident neutrons are detected. This is well in excess of the corresponding detection rate of prior art detectors.

Further, since the neutron interaction produces alpha particles, the signal in the detector will be very large; both the alpha particle and the lithium atom produced by the neutron's interaction with boron will travel very short distances (of order a few nm for Li, and a few  $\mu$ m for the alpha particle). Even over this short range, the products may produce in the region of 180,000 electron-hole pairs, depending on the incident neutron energy.

It may be desirable in some applications to reduce leakage by cooling the detector.

In one embodiment the boron-doping is restricted to a thin surface film. This may coat the upper surface of the ridges which may themselves be of intrinsic diamond. The coating may be of any suitable boron-rich substance such as borate. Preferably, the boron-rich coating should be thinner than the range of the emitted alpha particles, for example less than 20  $\mu$ m. In yet another alternative arrangement, boron may be layered in a sandwich structure within the ridges.

Boron-doped diamond as described above has a number of apparent applications, such as in monitoring devices, particularly in and around nuclear reactors and nuclear chemical plants (where it is essential to be warned of

the onset of accidental criticality). Further applications are envisaged in detection devices, such as analysis of radioactive waste, fissile material safeguards (where the level of radiation may be relatively low) and neutron thermopiles. The unusual properties of boron doped diamond make it particularly advantageous when used in the latter application, where the environment may be hot and hostile. In a reactor, the background from gamma radiation and other ionising particles may be removed by using a "double electrode" technique, or by simply gating the huge pulse produced by a neutron. The low cross section for gamma radiation interaction with diamond is therefore an advantage in the present case, and permits a counting rate and range substantially higher than that in known reactor power level monitors such as fissile detectors.

Boron-doped diamond also has substantial applications in diagnostic devices which detect and interrogate backscattered neutrons, for example in substances containing carbon or hydrogen. The backscattering medium acts in essence as a moderator, its low mass making scattering particularly effective since scattering depends exponentially upon the mass of the scatterer.

Thus by using the detector in combination with a rapid pulse source or an electronic chopper, it is possible to detect and interrogate drugs; this is possible because of the excellent time-of-flight and energy resolution capabilities of the detector. The high resolution and penetration capabilities also allow the detection of explosives and plastic explosives in particular, since they are constituted of materials having low atomic numbers. Finally backscattering of hydrocarbons may be interrogated, the thermal, radiological, chemical and

mechanical robustness of diamond being beneficial. Since the detector does not need a window, sensitive measurements down a bore-hole, for example, are possible.

5 The boron-doped diamond detector, being relatively compact, is especially suited to applications in the fields of continuous area monitoring and personal dosimetry. Small devices capable of detecting other forms of radiation are already known and can be incorporated  
10 with the boron doped diamond neutron detector. For example, to measure tissue dose over a wide range of energies, the detector must give an energy dependent dose response equivalent to that of human tissue. This may be done by using layers of polythene and screening materials  
15 together with the doped diamond.

The ridge-type detector with its spatial resolution (and the further embodiment with the perpendicular set of ridges which allows x-y positioning), together with the  
20 very rapid time response and high sensitivity, renders the detector suitable for novel applications in neutron radiography. In particular, it offers the possibility of neutron radiography of either static or dynamic systems containing moderating material of a smaller size and a  
25 more rapid dynamic response than has hitherto been possible. In neutron radiography, a parallel beam of neutrons impinges on the engineering component (containing some included moderating material). Any collision with this moderating material diverts the  
30 neutron from the parallel beam - effectively throwing an image of the system, highlighting the moderating material, upon a detector.

Although boron-doped diamond is of particular use in  
35 detecting slow neutrons, it will also detect fast

neutrons. For example, in neutron time and flight measurements, a fast response time together with high detection efficiency is required over all neutron energies. A number of planar boron-doped diamond detectors may be employed adjacent to one another, thereby increasing the area presented to the neutrons. This system improves upon known detectors such as lithium glass scintillators. By providing two diamond detectors, one of which is covered by a slow neutron filter such as a cadmium screen, it is possible to determine both the total number of neutrons incident as well as the respective amounts of slow and fast neutrons.

Another use is in neutron spectroscopy and neutron diffraction. A thin layer of material containing lithium or  $\text{He}^3$  might be placed between two diamond detectors. The reaction of the neutron with the lithium then produces a triton and an alpha particle. The fast detection rate of the diamond aids coincidence detection, whilst the summing of the two particle responses may be interpreted to yield the neutron energy.

Finally the use of the so-called "proton recoil" principle may also allow doped diamond to detect fast neutrons. A suitable proton-containing radiator would be used and the proton energy measured at a range of angles.

CLAIMS

1. A neutron detector comprising a plurality of boron-doped diamond detector elements having generally parallel sides, the sides carrying readout electrodes.  
5
2. A neutron detector as claimed in claim 1, wherein the boron concentration is  $10^{20}$  atoms  $\text{cm}^{-3}$  or less.  
10
3. A neutron detector as claimed in claim 2, wherein the boron concentration is  $10^{18}$  atoms  $\text{cm}^{-3}$  or less.
4. A neutron detector as claimed in claim 3, wherein the boron concentration is between  $10^{18}$  atoms  $\text{cm}^{-3}$  and  $10^{16}$  atoms  $\text{cm}^{-3}$ .  
15
5. A neutron detector as claimed in any preceding claim, wherein the detector element is formed by a growth method including chemical vapour deposition (CVD) or microwave or radio frequency plasma growth.  
20
6. A neutron detector as claimed in claim 5, wherein the boron is introduced to the diamond using diborane ( $\text{B}_2\text{H}_6$ ) gas or boric acid in acetone.  
25
7. A neutron detector as claimed in any one of claims 1 to 4, wherein the diamond is doped by ion implantation of boron into type IIA diamond.  
30
8. A neutron detector as claimed in claim 7, manufactured by an ion beam of concentration is  $10^{14}$  atoms  $\text{cm}^{-2}$ .
- 35 9. A neutron detector as claimed in any one of the



preceding claims in which the plurality of detector elements are formed from a single wafer of diamond.

- 5 10. A neutron detector as claimed in any one of the preceding claims in which the detector elements comprise non boron-doped diamond having a boron-rich coating thereon.
- 10 11. A neutron detector as claimed in claim 10 in which the coating is of borate.
- 15 12. A neutron detector as claimed in any one of claims 1 to 9 in which the detector elements comprise a sandwich structure of boron-doped and non boron-doped diamond.
- 20 13. A neutron detector as claimed in any one of the preceding claims in which the detector elements are mutually parallel, the space between adjacent elements being filled with an absorber material.
- 25 14. A neutron detector as claimed in any one of the preceding claims having a first detector element which is filtered by a slow-neutron filter, and a second exposed detector element.
- 30 15. A neutron detector as claimed in claim 11 in which the filter is of cadmium.
- 35 16. A neutron detector as claimed in any one of the preceding claims including an adjacent layer of a material containing lithium.
17. A neutron detector as claimed in any one of the preceding claims including an adjacent layer of a

material including He<sup>3</sup>.

5

18. A neutron detector as claimed in claim 13 or claim 814 in combination with a further neutron detector, the layer being sandwiched between the two detectors.
19. A neutron detector substantially as specifically described with reference to figure 2 or with reference to figures 3 and 4.

1 / 2

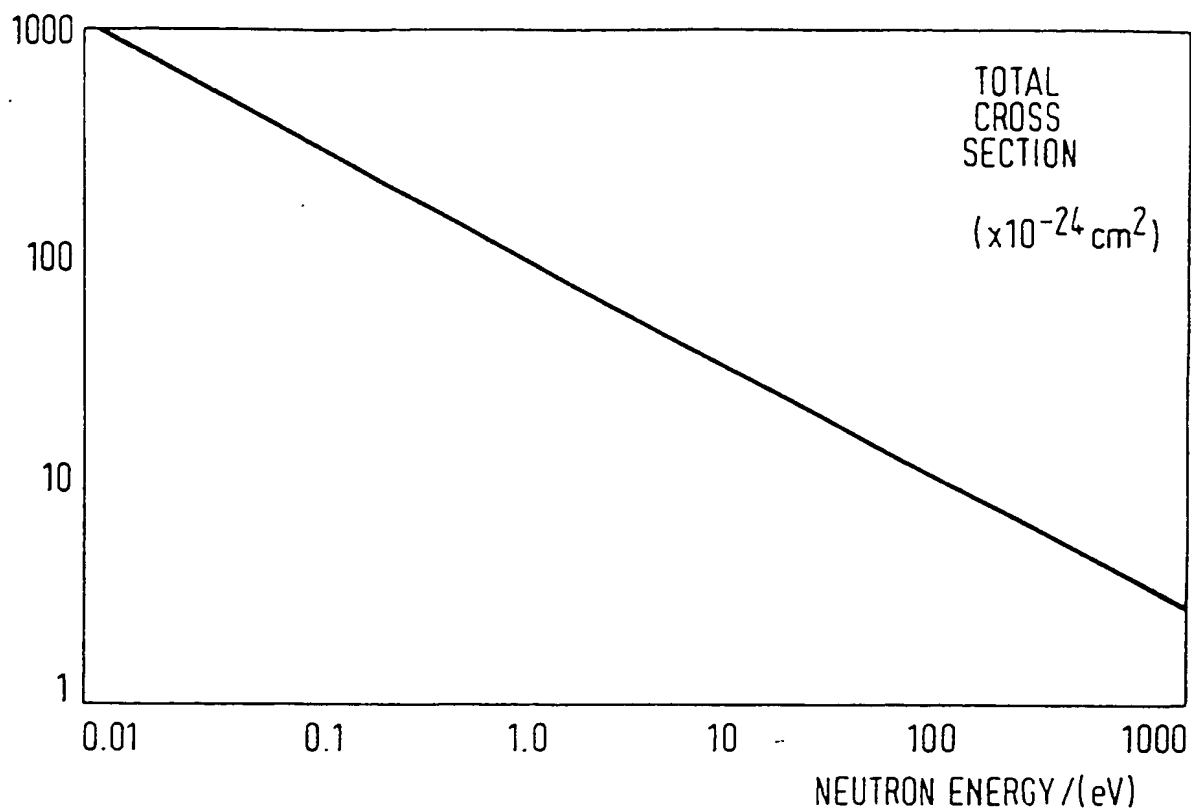


FIG. 1

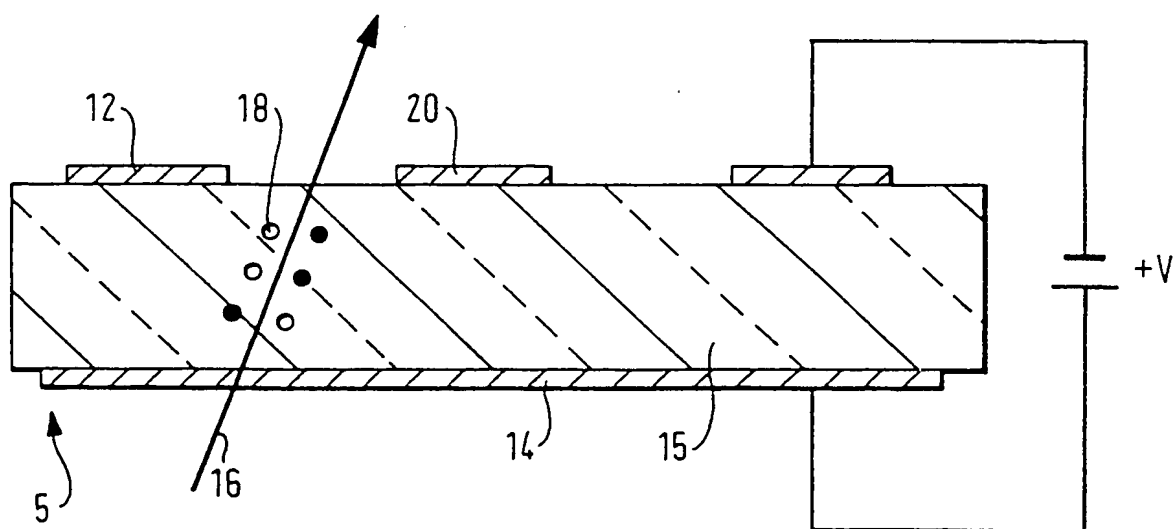


FIG. 2

2 / 2

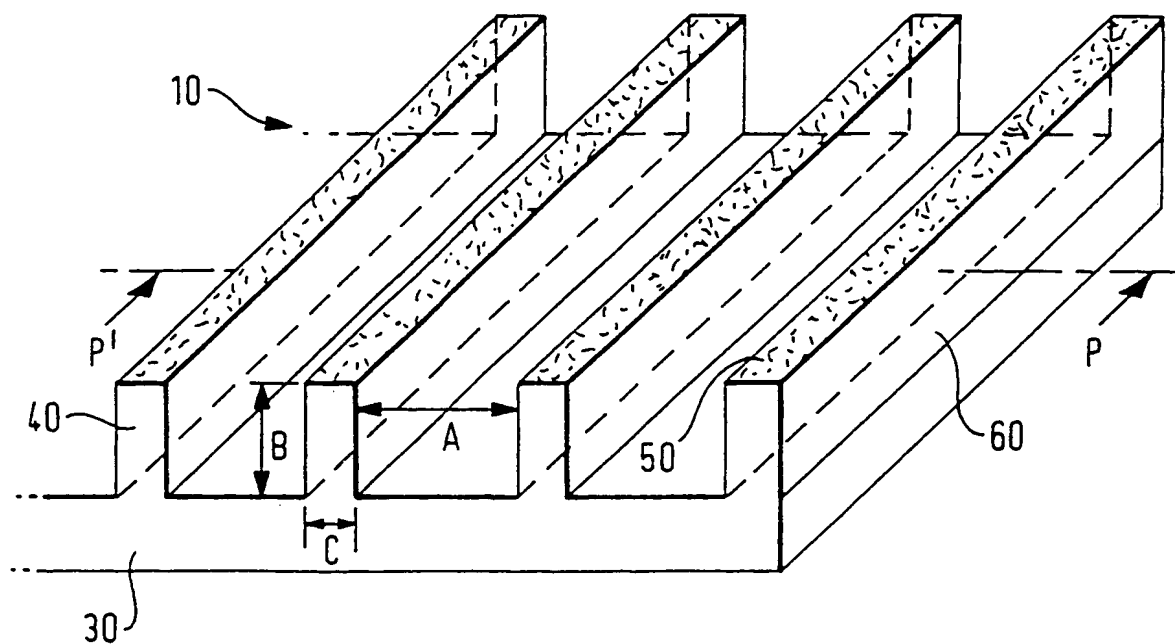


FIG. 3

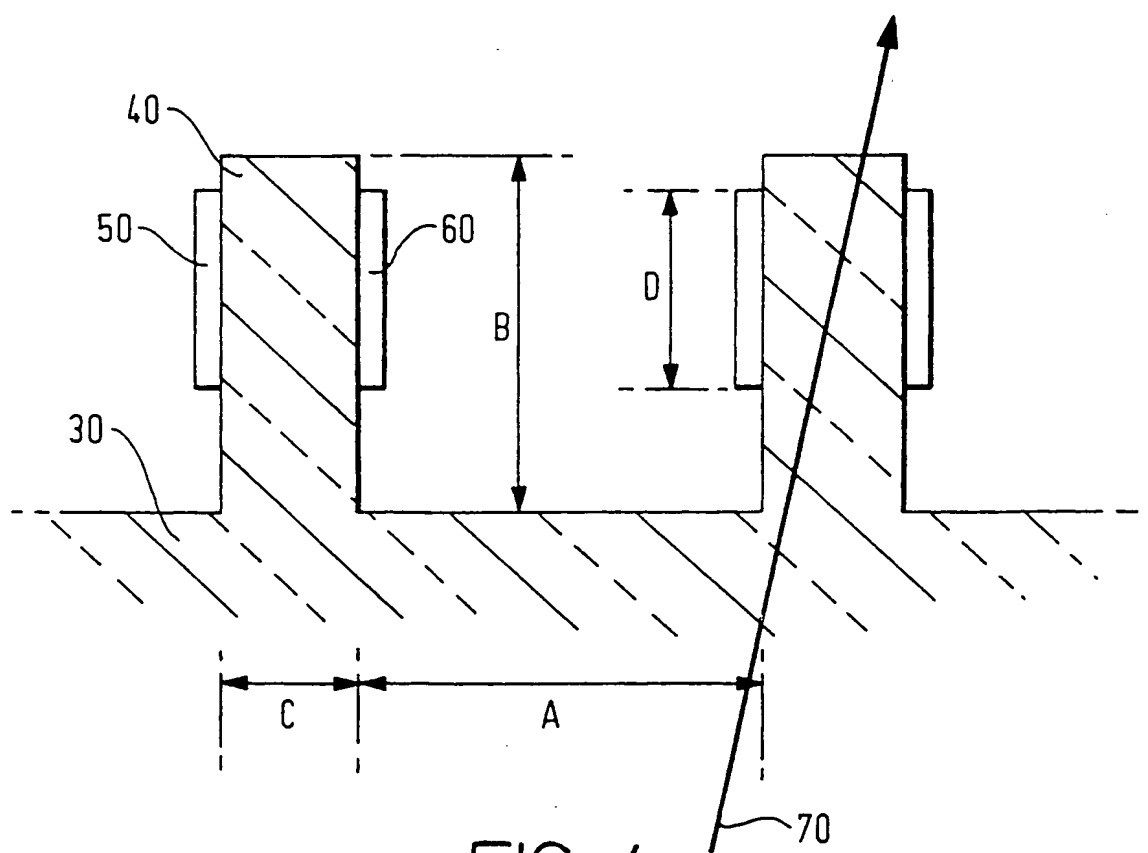


FIG. 4